From classical to quantum kisses between optical nanoantennas

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Abstract-A review of the recent capabilities of strongly-coupled optical gap-nanoantennas as building blocks to provide near-field coherent control, ultrafast all-optical switching and addressing high-frequency transport, among others, will be provided. As separation distances between metallic nanoantennas reach subnanometric dimensions, strong nonlocal effects are triggered out and coherent electron tunneling between the nanoantenna blocks is produced. A quantum-corrected model revealing the quantum regime in tunneling plasmonics is described.

Optical gap-nanoantennas formed by strongly-coupled metallic nanoparticles provide the capability to tune the optical response on demand. A switch between a capacitive and conductive situation within the gap modifies the plasmonic modes allowing for a variety of applications such as all-optical switching, near-field coherent control of single emitters, or probing of transport properties at optical frequencies. Most of these effects can be tackled within a classical treatment of electrodynamics since the typical distances involved in the optical interactions of plasmonic gaps are nanometric.

As metallic nanoparticles come close together at subnanometric distances, the plasmonic nanogap enters a strong non-local regime. For separations of ~0.35nm electron quantum tunnelling across the gap modifies dramatically the optical response of the system. To account for the effect of the spill-out of the electrons at the surface of the metal as well as the coherent tunnelling that can be established across the gap, full quantum mechanical calculations of the optical response are necessary. However, quantum-mechanical calculations can tackle a limited number of electrons effectively limiting the size of the nanostructures that can be addressed. Time-dependent density functional theory (TDDFT) has been successfully applied to obtain the response of a few thousand electrons, but the billions of electrons present in a realistic plasmonic structure exceeds the current capabilities of quantum frameworks [1,2].

We present a new method to calculate quantum effects in large plasmonic systems based on parametric inputs derived from simpler full mechanical calculations. With this quantum corrected model (QCM) [3], it is possible to obtain extinction cross sections as well as near-field enhancements for situations involving subnanometric interactions, thus bridging the gap between classical and quantum plasmonics.

Furthermore, the quantum-corrected model has been successfully applied to describe the tunnelling regime in an experimental situation where two metallic particles are located in subnanometric proximity, almost kissing each other [4]. A simultaneous measurement of the transport properties and the optical characterization by dark-field microscopy allows for capturing the quantum regime in tunnelling plasmonics by means of a sudden blue-shift of the plasmonic modes as the gap distance is decreased. Classical descriptions fail to address the modal distribution and the field enhancement in plasmonic gaps separated by less than ~5 Å. The presence of quantum tunnelling screens the charge densities induced at the gap and reduces the field enhancement establishing a fundamental quantum limit to the minimum volume of a metallic cavity where light can be trapped.

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